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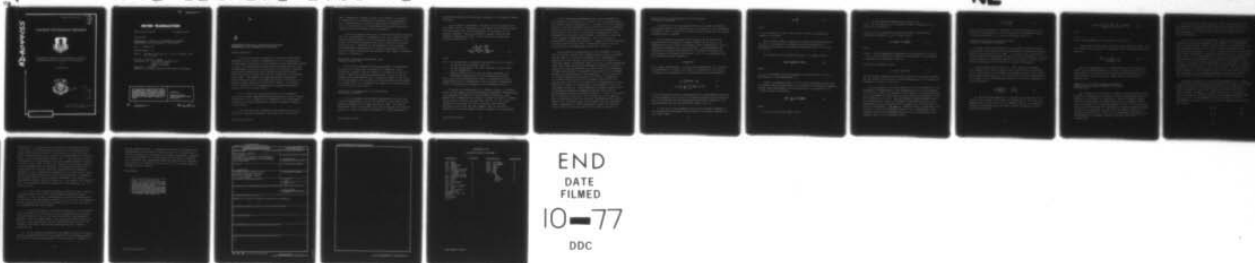
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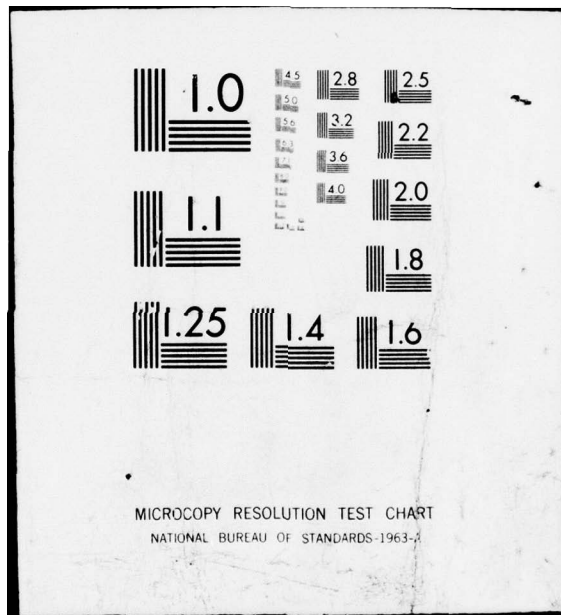
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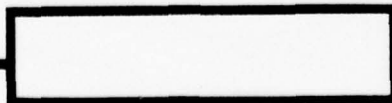
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## COMPARISON OF RESULTS OF ATMOSPHERIC POLLUTION MEASUREMENTS WITH STANDARDS OF AIR QUALITY

Andrzej Kasprzycki

In the article there is suggested a method of quantitative comparison of the results of measurements of the concentration of atmospheric pollutants with air quality, for an arbitrary interval of time. The quantitative criteria were determined on the basis of mathematical statistics by the method of confidence intervals. There were considered two basic variants; when the obtained results produced a simple test and when they produce a stationary time series. The problem of comparing emission measurement results with air quality standards was solved on the basis of an empirical model worked out in the USA by Larsen. The essence of the operation is the establishing of a guaranteed decision function for the two basic cases.

In the checking of the sanitary state of the atmosphere especially in urban agglomerations and industrial centers a highly significant role is played by the referring of the measurements obtained of pollutant emissions to legally establish standards of air quality.

In the case when the exceeding of the compulsory, for the given area, maximum permissible concentrations of pollutants

has a consequence a certain action, for instance in the form of a limitation of the emission of pollutants to the atmosphere, it is indispensable to establish quantitative criteria on the basis of which there will be undertaken the decision about such limitation, with a desired level of guarantee.

In the continuation there will be discussed problems connected with the establishment of a guaranteed (at a set level of confidence) decision function in the case where the standards of air quality are overrun and where they are not overrun at the given place and time. Similarly the consideration of two variants is indispensable namely where the obtained measurement results produce a simple test where they produce a time series of definite radius of correlation.

#### Processing of Emission Measurements that Produce a Simple Test

In the case of making checking measurements of atmospheric pollution in a given area in sufficiently large time steps (intervals), greater than the radius of correlation of the time series - among the successive measurements, or in the case of taking mobile measurements with a continuous change of the place of the instrument, the collection of data obtained can be handled as a simple test, i.e. accepting that there is no autocorrelation in the so-called time series.

#### Simple Test of Overrunning of the Established Air Quality Standards

It is established that emission measurements are done with a time of gathering(?)  $T_1$  and it is desired to refer them to air quality standards for the same interval of time. Let further the number (size) of the simple test (non-correlated time series) amount to  $N$  (and) according to assumption  $n$  ( $n \leq N$ ) measurements

exceed the established air quality standard  $Y_n$  of the given chemical substance.

In the example considered, a decision function guaranteed at an assumed level of confidence, is equivalent to the lower estimate from the random test of the probability of exceeding the air quality standard. Since the inequality  $y \geq Y_n$  occurs with an empirical probability  $p = \frac{n}{N}$ , if then there is satisfied the relation (See[1:§4.4]).

$$\frac{N}{N + t(b)} \left( p + \frac{t^2(b)}{2N} - \right. \\ \left. - t(b) \sqrt{\frac{p(1-p)}{N} + \frac{t^2(b)}{4N^2}} \right) \geq K \quad (1)$$

where

$t(b)$  - is the abscissa of Student's distribution for  $N-1$  degrees of freedom and confidence level  $1-b$ ,

$K$  - is the established in the given problem, critical value of the probability,

$y$  - random variable, in the problem being considered, the arbitrary concentration of the atmospheric pollutant,

Then on the given protected area there is undertaken an action conditioned on the overrunning of the air quality standard for a time of gathering  $T_1$  confirmed at the confidence level  $1-b$ .

At present in Poland there is in effect a manner of interpretation of the legally established maximum permissible concentrations of atmospheric pollutants, by some chemical substances, which permits the overrunning for instance of medium 24 hour(?) standards in the course of 5% of a day per year ( $K=0.5$ ), of a 20 minute standard in the course of 0.5% of a day per year ( $K=0.05$ ). Obviously in this case the test (sample) must be uniform in relation to the year's period, which in general is not satisfied.

In the established measurement place, for concentrations for instance of 20 minutes, it is best to use a so-called random test proportionately stratified [2]. In such a test the beginning element is selected randomly from the first 24 hours for measurement, the next from the 2nd 24 hours but now not randomly, it is shifted by a unit (that is by the time of gathering of a unit observation for instance  $T_1=20$  minutes, in relation to the time of the element already selected etc. The test can also be concentrated by taking for instance  $k$  elements at uniform distances from each other from the first 24 hour period and in the next 24 hour period all elements are shifted by a unit etc.

Such a manner of interpretation of the standards in effect for air quality, besides the incomprehensibility for the average user of them (i.e.) for one not making use of mathematical statistics) in the opinion of the author does not secure to a sufficient degree the hygienic state of the air in urban agglomerations since under critical meteorological conditions, so-called stagnations (lasting generally for several days), the appearance of high concentrations of atmospheric pollutants is not sporadic (random) but constitutes the rule generally. On the other hand it can also happen that the sum of the periods of stagnation in a particular place will be for instance less than 5% of a day per year, and then in the theory of the regulations in force, there are no cases of overrunning of the standards in force of atmospheric pollution. These regulations moreover ignore levels of atmospheric hazard in stagnation periods (each value of emission greater than the normative quantile of order for instance 5% is treated the same way). For information it is suggested that for instance in the USA there are standardized only maximum values (i. e. extreme values of the random test) for different times of gathering of a unit measurement, which (max values) can be overrun at the most once in the course of a year [3].

## Simple Test of not Overrunning of the Established Air Quality Standards

A situation of this type appears when on the basis of emission measurements made it is desired to have a guarantee (at the established confidence level  $1-b$ ) that at the given place and time the air quality standards are not overrun.

In the situation analyzed the conclusion has to do with the quantile which lies above the extreme element of the test; it is indispensable then to make an assumption which determines the type of statistical distribution of the population. For measurements of atmospheric pollution concentration it is possible to accept that they produce a log-normal distribution, i.e., it is postulated that the transformed random variable:

$$z = \log (y + C).$$

has a normal distribution. In the first approximation it is possible to assume that  $C = 0$  and on the basis of a random test (of size  $N$ ) to determine the parameters of the distribution from the equations:

$$m = \frac{1}{N} \log (y_1 y_2 \dots y_N) \quad (2)$$

$$s = \frac{1}{N-1} \sqrt{\sum_{i=1}^N \log^2 y_i - N \cdot m^2} \quad (3)$$

In the continuation there is indispensable the establishment the order  $K$  of the quantile  $y(K)$ , which in the analyzed statistical distribution can not be greater than the established air quality standard of the same gathering time as the measurements made, i.e.,  $T_1$ .

In the case of the assumption that the air quality standards in the contemplated measurement period cannot be in general overrun, we can accept (that):

$$K = \frac{T_1}{T} \quad (4)$$

where:

T - is the period of time in the course of which the random test (sample) is obtained.

When moreover there is permitted a certain established percent of time of overrunning the standard maximum concentrations of atmospheric pollutants, as already was explained above:  $K = 0.05$ ,  $K = 0.5$  etc.

On the basis of the assumptions made it is obvious (that):

$$z(K) = \log y(K) \text{ i } Z_n = \log Y_n \quad (5)$$

where:

$Y_n$  - is the standard value of atmospheric pollution (for a time of gathering of a unit measurement  $T_1$ ).

Further in agreement with [1: §4.7] with sufficient accuracy for the problem under consideration, it is possible to estimate the asymptotically normal dispersion of the distribution of the quantile  $z(K)$ , of order K from the equation:

$$S(K) = \frac{s}{\sqrt{N}} \sqrt{1 + 0,5 t^2(K)} \quad (6)$$

where:

N - is the size of the sample (test)

$s$  - is the magnitude determined by equation (3),  
 $t(K)$  - is the abscissa of the standard normal distribution for a value of its ordinate  $K$ ,

as well as in accordance with relatively simple considerations (specially discussed in [2]) if there occurs:

$$Z_n > s t(K) + m + s(K) t(a) \quad (7)$$

where:

$m$  and  $s$  - are the magnitudes determined by the equations (2) and (3)  
 $t(a)$  - is the abscissa of Student's distribution for  $N-1$  degrees of freedom and a confidence level  $1-a$ .

then at the confidence level:

$$1 - b = (1 - a) (1 - K) \quad (8)$$

one can accept (undertake the decision) that the air quality standards are not overrun (in the sense of the determination (4) of another for instance  $K = 0.05$ ;  $K = 0.5$  etc.).

It is necessary to note that equation (8) is a consequence of the assumption of independence of the determination (4) (or for instance  $K = 0.05$  etc.) and of the estimate of the quantile of the distribution (5) by the method of confidence intervals (with the dispersion (6) and the confidence level  $1-a$ ). Such an assumption appears natural since for instance in the case of knowledge of the precise values of the parameters  $m$  and  $s$ , in place of the inequality (7) it is possible to use the relationship:  $Z_n > s t(K) + m$ , reaching  $Z$  with a probability less than  $1-K$ . Since the confidence interval  $1-a$  is independent of the assumed course of the argumentation and it can be established arbitrarily, then if it is accepted (that):

$$1-a = \frac{1-b}{1-K} \quad (9)$$

then it will be possible to eliminate the auxiliary magnitude  $a$  and assure the interpretation of the results with the guarantee desired in the given problem, i.e., at the confidence level  $1-b$ .

#### Processing the Results of Emissions which Produce a Correlated Time Series

In many cases the acceptance of the assumption about the mutual independence of the results of the measurements of pollutants emissions leads to errors which are too large. In a case of this kind one has to deal with the interpretation of mean 24 hour concentrations of pollutants obtained in a continuous manner in the course of a year as also with the processing of the records of automatic instruments.

For stationary time series (i.e., which produce a dependent test (sample)) it is best to replace such a series with an equivalent to it, in the sense of the amount of information, simple test (sample) with a hypothetical size  $N_z$ . For this purpose there is indispensable an estimate of the autocorrelation matrix of the series determined by the relationship:

$$P = \begin{vmatrix} 1 & r_1 & r_2 & \dots & r_k \\ r_1 & 1 & r_1 & \dots & r_{k-1} \\ r_k & r_{k-1} & \dots & \dots & 1 \end{vmatrix} \quad (10)$$

In the first approximation it is possible to accept that the effective level (order) of the matrix is significantly less than the number (size) of the series (i.e.,  $k \ll N$ ) as well as to estimate the elements of the matrix with the help of point estimators from the general relationship

$$r_w = r_{l-j} = \frac{1}{s^2} \left( \frac{1}{N-w} \sum_{i=1}^{N-w} \sum_{j=1+w}^N z_i z_j - m^2 \right) \quad (11)$$

where:

$w = 1, 2, \dots, k$

$m$  and  $s$  are determined by the equations (2) and (2)

As was shown in the works [1] and [2] the substitute number (size) for a simple test equivalent to the given series is obtained from the equation:

$$N_z = \frac{N}{1 + \frac{2}{k} \sum_{i=1}^{k-1} (k-1)r_i} \quad (12)$$

The whole continuation of the argumentation is the same as for simple tests, whereby equations (1) and (7) remain proper in the continuation in the case of (by) replacing the number (size) of the actual series  $N$  by the substitute number (size  $N_z$  determined by equation (12).

Comparison of Pollution Emission Measurement  
Results with Air Quality Standards of Another  
Gathering Time

The concept of an air quality standard in protected areas refers generally to atmospheric air which is used by human groups (with the exception of factories) for breathing. Here the standard maximum possible concentrations of harmful substances of relatively short time of gathering of a single measurement (20 min. to 24 hours) must above all protect against the appearance of marked (sudden) instances of falling ill, moreover in order to insure against instances of becoming chronically ill, that is due to atmospheric pollution, the establishment of standards, monthly, seasonal, and yearly, - is indispensable.

The results of the view presented is that there exists a need for comparison of atmospheric pollutant emission measurement results, carried out in the region of their influence on people and plants, with standards of air quality with varied time-of-gathering of a single measurement.

In the case of a fixed atmospheric pollutant measuring network, supplied with continuous analyzers, for example of the coulometric type, the most rational manner of processing the measurement data for the above depicted goal, is the using of a yearly random sample, proportionately stratified. If such a sample establishes (constitutes) a population compounded of elements of gathering time  $T_1$ , for instance equal 20 min, it is indispensable to establish the relationships that make possible the determination of the population parameters (time series) of this same physical phenomenon. (i.e., atmospheric pollutions by a given substance) but of other times of gathering  $T_2$  of a single measurement, for instance one hour, 24 hours etc. For this purpose it is possible to use an empirical model of the distribution of the time intervals of the emission measurements suggested by Larsen [3 and 4] and based from the theoretical side on the treatment [2].

As earlier explained in the first approximation is possible to assume that the measurements of atmospheric pollutants on protected areas, of arbitrary time of gathering of a (single) measurement - produce log-normal statistical distributions (this means that the random variable  $z = \log y$  has a normal distribution). Keeping in mind that among the geometric mean  $m_g$  and the mean geometric deviation  $s_g$  as well as the parameters of the distribution  $m$  and  $s$ , determined by equations (2) and (3) there is reached (there occurs) the relation

$$m_g = e^m \quad (13)$$

$$s_g = e^s \quad (14)$$

on the basis of equations specially worked out in works [2] and [3] there is obtained:

$$m_2 = \ln \left[ m \left( \frac{\exp m_1}{m_2} \right)^w \right] \quad (15)$$

$$s_2 = w^{1/2} s_1 \quad (16)$$

$$w = \frac{\ln T/T_2}{\ln T/T_1} \quad (17)$$

where:

$m_1$  and  $m_2$  - are average values of the logarithmic distributions of the concentrations of atmospheric pollutants determined by equation (2) with time of gathering of a single measurement  $T_1$  and  $T_2$ ,

$s_1$  and  $s_2$  - are dispersions (stan devs?) of the logarithmic distributions of the atmospheric pollutant concentrations determined by equation (3) of time of gathering of a single measurement  $T_1$  and  $T_2$ ,  
 $m$  - is the average value of the distribution of atmospheric pollutants (of an arbitrary time of gathering of a single measurement and not logarithmic),

$T$  - base reference period of Larsen's time model, regularly one year (8760 hours).

In order to determine the decision function guaranteed at the confidence level 1-6, it is established that the hypothesis about the log-normal distribution of the pollutant emissions, as also the functions/relations (15), (16) and (17) are satisfied precisely. In such a case using the method of confidence intervals as well as the relationship (6) for a standard of air quality of time of gathering (accumulation)  $T_2$  there is then obtained:

- in the case when at the level of confidence 1-b the standards are overrun

$$\log Y_n(T_2) < s_2 t(K) + m_2 - t(a) \frac{s_2}{\sqrt{N}} \sqrt{1 + 0,5 t^2(K)} \quad (18)$$

- in the case when at the level of confidence 1-b the standards are not overrun:

$$\log Y_n(T_2) > s_2 t(K) + m_2 + t(a) \frac{s_2}{\sqrt{N}} \sqrt{1 + 0,5 t^2(K)} \quad (19)$$

where:

$N$  - is the number (size) of the original time series (of time of gathering of a single observation  $T_1$ ),

$m_2$  and  $s_2$  - are the magnitudes determined by the relations (15) and (16) for the point estimators (2) and (3) of a time of gathering  $T_1$  of a single observation  $y_i$ ;  $i = 1, 2 \dots N$ .

$Y_n(T_2)$  - are the standard's maximum concentrations of atmospheric pollutants, average for the time interval  $T_2$ ,

$t(a)$  and  $t(K)$  - are quantities of the same significance as in (7) and (6).

## Conclusions

On the basis of the presented considerations it is possible to formulate the following more important affirmations, which can have practical applications in the control of the sanitary state of the atmosphere in large urban agglomerations.

1. Before approaching the given protected area for active influencing of the magnitude of pollutant emissions to the atmosphere, as also during the process itself of establishing permissible values of

emissions, it is indispensable to introduce quantitative criteria, on the basis of which tactical decisions will be undertaken (this means short term limitations of the emissions by some industrial plants, implementation of the process of desulphurization of chimney gases in electric generating plants, etc.). Such criteria are also indispensable for determining of the joint maximum amount of days in the year in the course of which the reduction of pollutants emitted to the atmosphere will be necessary. It seems that in the case of accumulation of a suitably broad and representative measurement material, of the concentrations appearing on the given area of atmospheric pollutants (emissions), the most rational way of comparing the obtained results with air quality standards is the method of confidence intervals, which introduces as a consequence the establishment of quantitative criteria, expressed by the equations (1), (7), (18), and (19).

2. In the case of ordering (giving instructions for) a rather long time series (this means a chronologically obtained random sample) it is best to use a proportionally stratified random sample as well as to use quantitative criteria (1) in the case of overrunning of the established air quality standards and (7) in the case where the standard has not been overrun.

3. In case of necessity of maximum exploitation of the information contained in the time series, it is indispensable to estimate the order (number, size) of the substituted series (determined by equation (12)), and right after to use the quantitative criteria (1) and (7). Such calculations can get effectively done only with the support of electronic calculating technology which is today in universal use.

4. In the control (checking) of the hygienic state of the atmosphere an important matter is the comparison of the obtained measurement results (emission measurements) with air quality standards of

various gathering times. In applications there is recommended the experimentally tested model of Larsen as well as the quantitative criteria (obtained by the method of confidence intervals) expressed by the relationship (18) in case where the air quality standards (of a different unit measurement gathering time than the original time series) are overrun, as well as by (19) in the case where the standards are not overrun.

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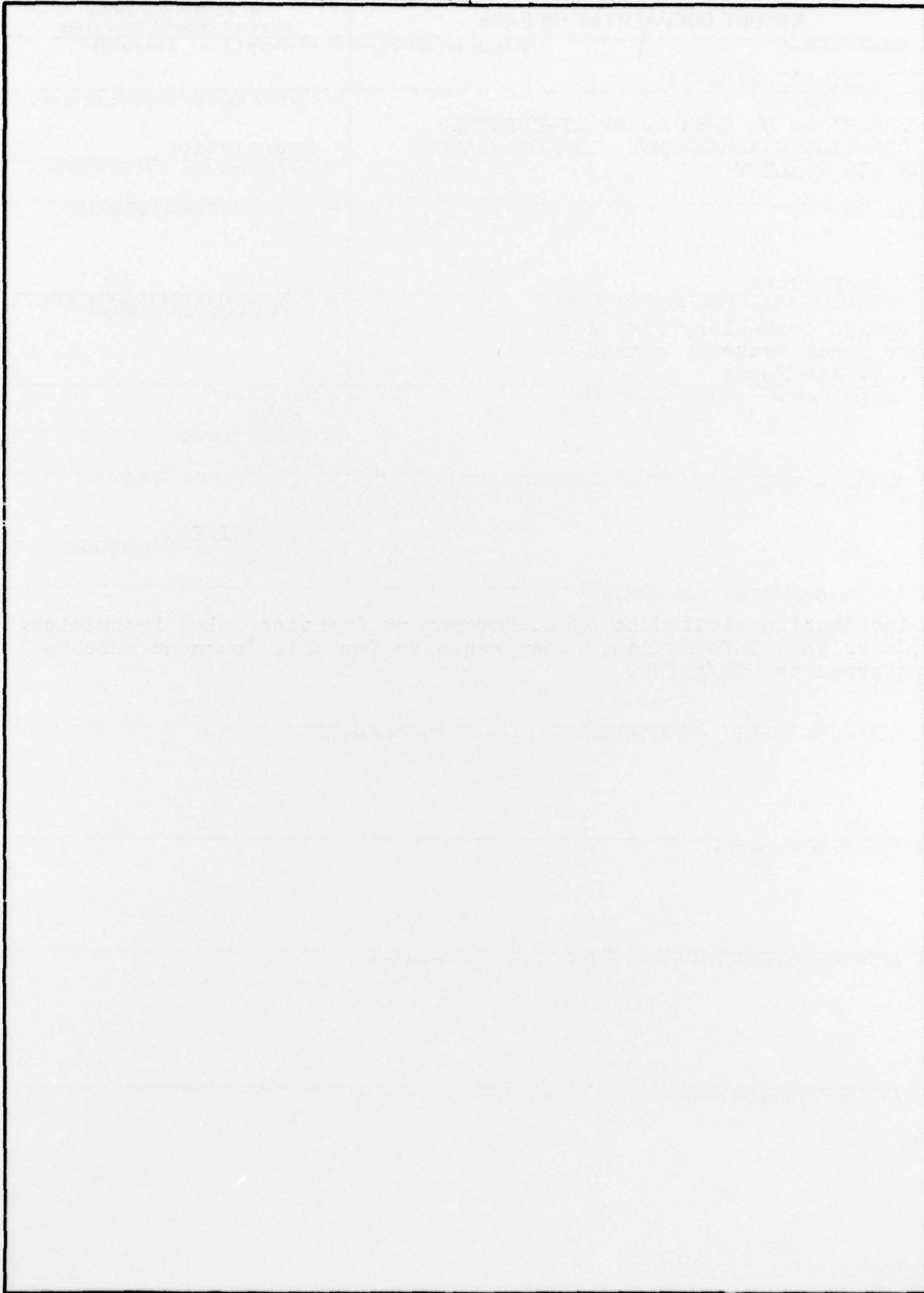
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